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# Gamma irradiation effects on the properties of indium zinc oxide thin films

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ABSTRACT

Thin films of amorphous and polycrystalline indium zinc oxide (IZO) having various In/(In + Zn) nominal compositions from 0.3 to 0.9 were grown by the pulsed laser deposition technique at room temperature or at 400 °C on oxidized Si and quartz substrates. After deposition, the films were gamma irradiated at doses of 10, 20 and 30 kGy under Ar atmosphere. Grazing incidence X-ray diffraction investigations could not find measurable changes of the structure after irradiation. Simulations of the X-ray reflectivity (XRR) and diffuse scattering curves acquired from the films showed a small density decrease after irradiation, accompanied by an increase of the thickness and surface roughness. Spectroscopic ellipsometry investigations confirmed the increase of the IZO films' thickness after irradiation, being in agreement with the XRR results. Small changes in the refractive index, most probably caused by changes in the film density were also observed. Four point probe measurements showed very small resistivity increases after irradiation, with the films containing higher In concentration presenting negligible changes.



## 1. Introduction

Amorphous oxide thin-film transistors (TFTs) have been extensively studied in the last decade due to their better optical and electrical properties compared to amorphous-silicon TFTs [1–4]. A major advantage for the amorphous oxide films is the fact that they could be manufactured at low processing temperatures, from room temperature up to 150 °C, on inexpensive, light weight and flexible polymer substrates. Such light structures make the amorphous oxide based TFTs attractive for space applications. There have been few studies so far about the effect of radiation on their performance and reliability, which is nevertheless of crucial importance as space materials are submitted to ionizing radiation. The purpose of the present paper is then to investigate how gamma radiation could affect the properties of such materials.

Jeon et al. [5] investigated the effect electron-beam irradiation on the indium zinc oxide (IZO) properties and found out that the electrical conductivity was increased after irradiation. Liu et al. [6] studied the effect of ionizing radiation on the IZO TFTs. After irradiation, a negative shift of the threshold voltage, accompanied by increases of the electron field effect mobility, subthreshold swing, and low frequency noise were observed. The effect of gamma radiation on the TFTs characteristics has been recently investigated by Indluru et al. [7]. The authors reported an increase in electron mobility from 2.8 to 8.8 $cm^2/Vs$ after the exposure, which was attributed to the combined effects of interface states creation and electron–hole pair generation in the insulating layer.

We deposited amorphous IZO thin films having different In/(In + Zn) compositions by the pulsed laser deposition (PLD) technique [8–10] to investigate their optical and electrical properties, structure and composition. The results showed that the deposited IZO films exhibited good optical properties in the visible and near infrared range, low resistivity and high mobility [8]. The carriers' mobility estimated from Hall and optical reflectometry measurements showed no significant differences, as expected for an amorphous material, where grain boundary scattering should not be present [10,11].

In this article we investigated the effect of gamma radiation on the structure and on the optical and electrical properties of amorphous and polycrystalline IZO films having In/(In + Zn) compositions in the 0.3–0.9 range, where the best combination of optical and electrical properties for TFTs has been previously observed [8,12,13].

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## 2. Experimental details

A KrF* laser source (model COMPexPro 205, Lambda Physics-Coherent, $\lambda = 248$ nm and $\tau_{las} = 25$ ns) was used to ablate homemade IZO targets with nominal In/(In + Zn) ratio concentrations from 0.30 to 0.90. An optimum oxygen atmosphere of 1 Pa was used during ablation, as determined during previous growth studies [6,7]. The laser fluence onto the target surface was around 2 J/cm$^2$ at a repetition rate of 10 Hz. Since we were interested in obtaining amorphous films, the quartz and oxidized Si substrates were not intentionally heated during the deposition. A thermocouple attached to the substrate heater indicated that during deposition the temperature increases by 5 to 10 °C. To investigate the effect of crystallinity on the properties of irradiated films, a series of samples were also deposited at 400 °C. After deposition, some of the films were submitted to gamma radiation from a $^{137}$Cs source (Gammacell 3000 from Nordion) at a dose of 10, 20 or 30 kGy (1 Gy = 1 J/kg) at a dose rate of 5 Gy/min. Prior to irradiation the samples were sealed in glass ampoules under vacuum to remove any dioxygen molecules that could interfere with the results and to mimic the space conditions. To accurately measure the changes induced by irradiation, each sample was measured before and after irradiation. There were some small variations of the initial thickness of the samples used for the study, which did not affect the results or conclusions since we monitored the relative changes.

The structure of the deposited films was investigated by Grazing Incidence X-Ray Diffraction (GIXRD) with the help of an instrument (Empyrean from Panalytical, Cu Kα radiation) working in a parallel beam geometry. The films' mass density, thickness and surface roughness values were obtained from simulations performed using a commercially available software (X'Pert Reflectivity) of X-ray reflectivity (XRR) and diffuse scattering curves acquired using the same instrument.

The thickness and refractive index values at 630 nm of the thin films resulted from optical analyses of ellipsometry spectra acquired with a Woollam Vertical–Variable Angle Ellipsometer (V-VASE) [14]. The ellipsometer was equipped with a HS-190 monochromator which enabled the spectral inspection of thin films. The elemental composition of the films was determined by means of Rutherford Backscattering Spectrometry (RBS) analysis performed with $^4$He ions at 2.6 MeV. The measurements were performed using a $^4He^{2+}$ beam extracted from the Alphatross ion source of the 3 MV Tandetron accelerator of HH-NIPNE. The alpha particles were detected with a passivated, ion implanted silicon detector placed at 165° with respect to the incident beam direction. The solid angle subtended by the detector was found to be 1.641 msr, while the energy resolution was 16 keV. The acquired spectra were simulated using the RUMP program [15,16]. The chemical bonding was investigated by X-ray photoelectron spectroscopy (XPS) with a Physical Electronics PHI 5100 instrument (Al Kα radiation, $h\nu = 1486.6$ eV, 300 W). The electron pass energy was 35.75 eV and the charge correction of the binding energy was performed using the known position of the C-(C,H) line in the C 1s spectra at 284.5 eV. The XPS spectrometer was calibrated using a polycrystalline Au foil. The peak position for the Au f7/2 peak and Fermi-edge inflection point were determined to be 84.00 ± 0.002 and 0.00 ± 0.02 eV, respectively. The core-level peaks were fitted using Voigt functions, after a Shirley-type background was removed from the acquired data. The sheet resistance of the deposited films was measured at room temperature under atmosphere and day light with a home-build in-line four point probe system (a nanovoltmeter – 2882 A, current source 6220 – from Keithley and a head probe from Jandel).

## 3. Results and discussion

The In/(In + Zn) averaged values measured by RBS on several locations on the films' center (an area of approx. 1 cm$^2$) are displayed in Table 1. The composition of the films was slightly In rich as compared to the composition of the target, probably a result of Zn higher partial vapor pressure than In in the laser plume. However, in order to facilitate the comparison of results we maintained the initial labeling of the samples that have been used in several already published articles [6,7]. A typical XPS survey spectrum acquired from an IZO_70 film is displayed in Fig. 1. The C 1s signal comes from a thin (~1–2 nm) contamination layer, which formed when the sample was exposed to the ambient. Also, the O 1s peak, displayed in Fig. 2, is a convolution of three components corresponding to metal–oxygen bonds (M–O, 530.2 eV), oxygen near oxygen vacancies (M–OV 531.5 eV), and oxygen bonded in a metal hydroxide (M–OH 532.6 eV) [17,18].

**Table 1**
RBS analysis of the average composition of deposited IZO thin films.

| Sample label | In/(In + Zn) |
|---|---|
| IZO_30 | 0.35 |
| IZO_50 | 0.57 |
| IZO_60 | 0.64 |
| IZO_70 | 0.79 |
| IZO_90 | 0.93 |

The relative intensity of the M–OH peak strongly decreased when the takeoff angle was changed from 45° (more surface sensitive) to 90° (more bulk sensitive), suggesting that the hydroxide signal corresponds to a very superficial layer present on the surface. Both the adventitious C 1s and hydroxide O 1s peaks disappeared after 5 min Ar ion sputtering.

GIXRD patterns displayed in Fig. 3 show that the films deposited at room temperature are amorphous. Some short order still exists, since the broad and asymmetric peaks can be easily fitted by two peaks located at 2θ values that would correspond to the strongest diffraction lines of hexagonal ZnO (ICDD Reference code: 98-002-6170, $d_{(002)} = 2.6035$ Å, $2\theta = 34.420°$) and bixbyite $In_2O_3$ (ICDD Reference code: 00–006-0416, $d_{(222)} = 2.9205$ Å, $2\theta = 30.586°$). This is in agreement with recent investigations showing that the In–O and Zn–O atomic distances in amorphous films are similar to that measured for crystalline ones [19]. After irradiation, there were no measurable changes in the diffraction patterns, as shown in Fig. 3b, where the patterns acquired from the IZO_90 films before and after irradiation at a dose of 20 kGy are displayed. In Fig. 4 there are displayed GIXRD patterns acquired from an IZO_90 film deposited at 400 °C. The film is crystalline, with average crystallite size of around 16 nm and a micro-strain of $3 \times 10^{-3}$. The pattern collected after an irradiation at a dose of 30 kGy and also displayed in Fig. 4 is very similar to the initial one. Films having lower In concentrations deposited at 400 °C were still amorphous, confirming

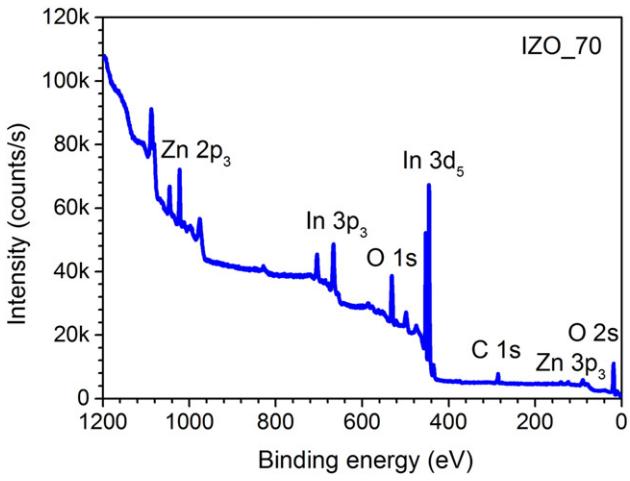


**Fig. 1.** XPS survey spectrum acquired at a 45° take off angle from an as-deposited IZO_70 film.